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Figure S1. Schematic representation of probable interaction mode between anionic amphiphiles 1-5 and representative hemicyanines. It is suggested that β -Ala residues of 1 and GABA residues of 5 are not directly responsible for dye incorporation because electrostatically bound dyes do not reach the β -Ala and GABA moieties of 1 and 5, respectively, and they rather play crucial role in adjusting packing mode between amphiphiles for formation of specific supramolecular environments (*i.e.*, width and hydrophobicity of cavities) whether they can incorporate hemicyanine dyes specifically or not.

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Figure S2. (a) UV-Vis. absorption spectra of aqueous solutions of St-4C₁ in the presence of **10**, **11** and **12**: temperature 20°C, pH 10, path length: 0.1 cm, [St-4C₁] = 0.15 mM = const., [**10**] = [**11**] = [**12**] = 3.0 mM: (b) Relationships between λ_{max} of St-4C₁ and spacer methylene number of L-Lys-derived amphiphiles **10**, **11** and **12**.

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(a)

pH2.6	pH3.2	pH3.8	pH5.1	pH6.1	pH7.2	pH8.0	pH9.1	pH10.1	pH10.8
2(C8-0410)-145 0A12-04/32-4C 042.8	1(21-AAla)-LLys- Adle-CHI / SC-HC1 H 3.2	21(0+.0A10)-1.Lys 0414-04/25C-42 943.8	21(ca-AAla)-Liys- A4la- 04/St-26C pH 3.1	2(Co-13ala)-145- Aala-04/55-46. pH 6.1	3((q. AAla)-ily5 AAla-0H/St-40 pH 7.2	2(Ca-AALA)-LIJS- BAIA-OH/SE-4- PH 2.2	31(20-00.4163)-1155" 0.41.0-04 / Se-46 pH 9. J	2(Ca- AAla)-145 AAla-CH / St-4C 541/0,1	2(Ca-AAla)-Ilys AAla-OH/St-44: PHIE.8
								M	

(b)

pH3.8	pH5.1	pH6.1	pH7.2	pH8.0	pH9.1	pH10.1	pH10.8
SIGE-MAIN)-LLYS- MIA-OH/SE-DC: PH 3.8	aller-Anta)-ilys- Arte-ont/St-42- pH S.1	2100+19410)-1145- 19814-04//St-40, pH 6.1	2(Co-AAIa)-1145- AAIa-CH/St-40- pH 7.2	2 (58-AA 14)-dys- BAta-CH/ 55-46 pH 8.0	31(ca-dala)-1105 AAla-04/30-40- ph 9, j	3(64-1941a)-1845- 1941a-047 St-186- 19470.1	2100 - Malal-Ulf Mala-CH/ 32-4- PHIR. 8
	-		-	-	-	-	-
	рН3.8 ^{310:040)-1,3*} ^{Milloy1,20:00} ун з.8	pH3.8 pH5.1	pH3.8 pH5.1 pH6.1	pH3.8 pH5.1 pH6.1 pH7.2	pH3.8 pH5.1 pH6.1 pH7.2 pH8.0	pH3.8 pH5.1 pH6.1 pH7.2 pH8.0 pH9.1	pH3.8 pH5.1 pH6.1 pH7.2 pH8.0 pH9.1 pH10.1

(C)

pH1.6	pH2.0	pH2.5	pH2.9	pH3.2	pH3.5	pH4.0	pH4.6	pH5.4	pH10.8
St-4C, : pH1.6	St-4c, : pH2.0	St-4C, : pH2.5	St-4C1 : pH2.9	St-4C1: pH3.2	St-4C, : pH3.5	St-4C, : pH 4.1	St-4C,:pH±6	St-4C, : pHSt	St-4C1 : pHIN.
				-				-	
3	1 3	-		-	-	and a strength of the			-

(d)										
	pH1.6	pH2.0	pH2.5	pH2.9	pH3.2	pH3.5	pH4.0	pH4.6	pH5.4	pH10.8
	St-4c, : pH1.6	St-4c.: pH2.0	St-4C, : pH2.5	St-4c1 : pH2.9	St-4C1: pH3.2	St-4C, : pH3.5	St-4C, : pH 4.0	St-4C, : pH 26	St-4c, :pHS?	St-4C1 : pHIO.
										_

Figure S5. Relationships between pH and the coluor of 0.15 mM of St-4C₁ in water in the presence of 3.0 mM of amphiphile **15**; (a) without black light irradiation; (b) under black light irradiation; (c) St-4C₁ alone in water without black light irradiation; (d) St-4C₁ alone in water under black light irradiation.

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Figure S4. Schematic illustration of pH-dependent expansion and contraction behaviours of L-lysinederived amphiphile 15 with long side-chain and L-glutamic acid-derived amphiphile 9 with shorter side-chain. Red and orange colours denote the pH-dependent colour change in St-4C₁ incorporated in self-assemblies of 15 and 9. St-4C₁ electrostatically bound to carboxylate of adjacent amphiphile is omitted for clarity because of [amphiphile]/[St-4C₁] = 20.

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Figure S5. Transmisstion electron micrographs of peptide amphiphile **15** (a) alone in water, and after addition of (b) LiCl and (c) NaCl; pH 10, [**15**] = 3.0 mM, [LiCl] = [NaCl] = 30 mM.



Figure S6. Representive DSC thermograms of peptide amphiphile **15** before (a) and after (b) addition of NaCl; pH 10, [15] = 20 mM, [NaCl] = 200 mM, heating rate: 2 °C/min.

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